2nd Quarterly Report
Period of 10 July to 10 October 1962

FUEL CELL ASSEMBLIES

Prepared for

Jet Propulsion Laboratory
California Institute of Technology
4800 Oak Grove Drive
Pasadena, California

Attn: Mr. J. McMahon
Contract No. JPL 950258

EOS Report 3070-Q-2 15 October 1962

Prepared by

Harvey Frank

Harvey Frank
Project Supervisor

Approved by

B. M. Wilner, Manager
Chemical and Fluid
Systems Department

Approved by

J. Neustein, Manager
ADVANCED POWER SYSTEMS DIVIS
DIVISION

ELECTRO-OPTICAL SYSTEMS, INC. - PASADENA, CALIFORNIA

TABLE OF CONTENTS

LIST OF FIGURES

1. SUMMARY

18492

The purpose of this program is to design, develop, and then deliver three multicell regenerative hydrogen-oxygen fuel cell assemblies to the Jet Propulsion Laboratories, Pasadena, California. The work is being carried out under JPL Contract 950258 (Fuel Cell Assemblies). 18492

The major accomplishments during the first quarter consisted of designing and beginning fabrication of the first multi-cell unit. Fabrication techniques and test procedures were established for all component parts.

This report describes the results of all investigations conducted on this program for the three month period from 15 July to 15 October 1962.

The major portion of the experimental activities during this period consisted of completing the fabrication, assembly, and initial testing of the first multi-cell unit regenerative hydrogen-oxygen fuel cell. The tests consisted of measuring the voltage-current and discharge characteristics for short cycles, internal temperatures, leak tightness, and capacity. Related fuel cell investigations were concerned with methods for increasing cell capacity, and optimization of catalyst. Materials studies were concerned primarily with evaluating high temperature plastics for the cell spacers.

Analytical activities during this period consisted of evaluating the potential safety hazards that may exist in the operation and handling of this type of cell. Also included were design calculations and preliminary planning for the development of the final units.

The work remaining on this program consists of designing, fabricating, and then testing the final units. In order to meet the delivery date of 10 February 1963, it will be necessary to complete the design and then begin fabrication of these units in the early portion of the next quarter.

2. MULTI-CELL UNIT TESTING

The following section contains the results of tests conducted on the first multi-cell unit. A brief description of the testing procedures as well as an illustration of the full scale assembly are given in the first subsection.

2.1 Testing Procedures

Two modes of operation are possible with the first multi-cell unit (see Fig. 1). In the first mode, the gas cylinders are removed from the assembly and the cell stack is run in a "quasi-secondary battery" type of operation, i.e., during charge, the gases formed by electrolysis are vented to the atmosphere and during discharge, the gases are supplied to the cells from externally located hydrogen and oxygen tanks. This mode of operation permits the testing of each individual cell within the stack. The unit is placed on load and the voltage probes are made of each individual cell.

In the second mode of operation, the gas cylinders are assembled to the cell stack and the assembly is run as a true secondary battery. In this mode of operation, the gases formed during charge by electrolysis are stored within the sealed assembly. On discharge, the same gases are returned to the cells.

During the report period, the major portion of the tests were conducted by the first mode of operation described above. Near the end of the report period when the functionality of each individual cell within the stack had been established, the unit was sealed with the gas cylinders and run as a true secondary battery.

Both charge and discharge tests were conducted at constant current. The charging current was supplied by a large constant current power supply. The unit was discharged through a 100 ohm variable

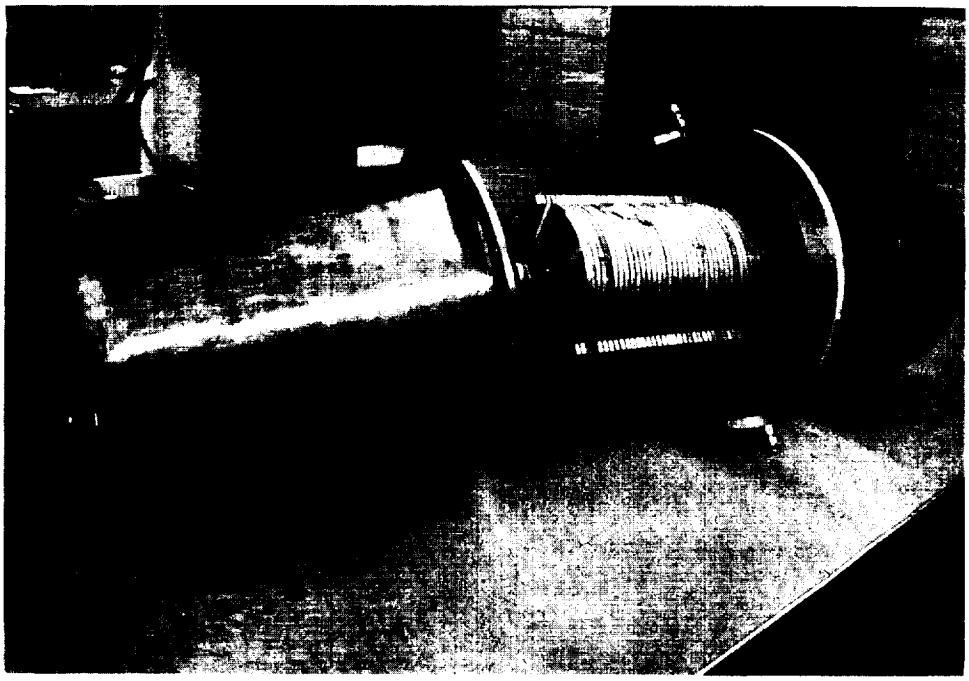


FIG. 1 FULL SCALE MULTI-CELL ASSEMBLY

rheostat. The discharge current was held constant by manual control of the rheostat.

2.2 Voltage-Current Characteristics

The following data was obtained by the first mode of operation described above. The first set of data gives the discharge voltage-current characteristics for two cells at 50 psig, room temperature, and containing electrodes prepared by the latest process (see Section 3.1).

Volts	Amps
2.15	0
1.75	5
1.52	10
1.20	15
0.82	20

The following data was for 30 cells at 5 psig, room temperature, and with the same electrodes as above;

Volts	Amps
30.5	0
25.0	1

Due to the low gas pressures, stable operation at currents greater than 1.0 amp could not be maintained, i.e., the voltage began to decline during the conduct of the test. This latter result signified gas starvation. Operation at higher pressures with a large stack of cells can only be carried out when the unit is completely sealed and run as a secondary. Since the latter mode of operation has only recently been initiated, there was insufficient time before the writing of this report to measure the voltage-current characteristics at higher pressures. These tests, however, are currently in progress and the results will be reported as soon as possible.

2.3 Capacity

Preliminary tests have indicated that the maximum capacity of the existing configuration is between 20 and 25 amp-hrs. The tests consisted of cycling multiple-cell assemblies according to the first mode of operation at successively higher inputs, i.e., 5, 10, 15 amp-hrs, etc. After an

input of 25 amp-hrs, the subassembly could be discharged; however, the performance was found to be very sensitive to pressure differential, i.e., a slight increase in pressure differential ($>$ 1 psi) between H_2 and O_2 caused the voltage to fall rapidly to zero. The results indicate that a charge input in excess of 25 amp-hrs may deplete the water content of the asbestos bed to such a degree that the bed may not be capable of effectively separating the gases. When operating in the true secondary mode, the gas pressures are balanced by a compensating bellows, hence, the previously described problem may not be encountered. However, if additional capacity is desired on the final units, one or more of the following modifications may be employed:

1. increase the diameter of the electrolyte bed
2. add an additional electrolyte layer
3. employ a lower KOH concentration

The feasibility of the second method is currently being examined on small experimental cells.

2.4 Ten Ampere-Hour Cycle

The 30-cell assembly was run in the first mode of operation for several short cycles between 0 and 2.0 amp hrs. The longest cycle which was conducted on this assembly was 10.0 amp-hrs. In this test, the assembly was charged at 4.0 amps for 2.5 hrs and was then discharged at 1.0 amp for 10 hrs. The charge and discharge voltages were 56 volts and 25 volts respectively. The discharge period was not continuous but was intermittent over a two-day period.

2.5 Experimental Heat Transfer Data

Experimental heat transfer data was obtained by instrumenting the middle cell of a stack of five cells with thermocouples and then measuring temperature vs. time on discharge. Two of the thermocouples were in contact with the inside edges of the hydrogen and oxygen electrodes respectively, and a third was in contact with the outside edge of the cell spacer. The stack of cells was exposed to the air and therefore, dissipated heat by natural convection.

Two results were obtained from this test. The first result was that the temperatures of both H_2 and O_2 electrodes were within $1^{\circ}F$ of each other at all times during the test. The second result was that under steady state conditions, i.e., constant temperatures with respect to time, the temperature drop between the inside edge of the electrodes and the outside edge of the spacers were found to be approximately $8^{\circ}F$ and $10^{\circ}F$ for electrical loads which would correspond to 94 watts and 162 watts respectively for a bank of 38 cells. The theoretical temperature drops for the above loads as indicated in the first quarterly report were $3^{\circ}F$ and $8^{\circ}F$ respectively. These results indicate a relatively small contact resistance between the electrodes and the cell spacer.

2.6 Full Scale Secondary Operation

When the functionality of each of the cells within the stack of 34 cells had been established, the gas cylinders were attached to the assembly and the unit was run as a true secondary battery, as described above.

The first tests to be conducted in this manner consisted of several short cycles of 2 amp-hrs. The units were charged at 2 amps for one hour and then discharged at 1.0 amp until the voltage began to fall rapidly to zero. In the first two cycles, the voltage began to decline in less than one hour. Subsequently, after having purged the gas cylinders more thoroughly, the voltage held constant for much longer periods of time (approaching two hours). The results indicated that thorough purging of the cylinder before final assembly is a very essential operating procedure for the removal of residual air.

A 10 amp-hour cycle is currently in progress and a 20 amp-hr cycle is scheduled thereafter.

3. FUEL CELL INVESTIGATIONS

This section contains the results of several investigations concerned with improving the performance of the fuel cell portion of the assembly.

3.1 Electrode Fabrication

The results of a related fuel cell program within EOS (NAS 7-181) indicated that a significant increase in electrode activity can be obtained by a modification of the platinization procedures. Exploratory runs with the electrodes prepared by this new process were therefore carried out and it was established that higher currents can be obtained with less polarization. Therefore, the decision was made to adopt the process for the preparation of all subsequent electrodes.

The main difference between the old and the new process is that of applying the platinum to the porous nickel. In the old process the porous nickel was merely agitated in a platinum chloride solution while in the new process the solution is forced through the porous nickel by means of a vacuum pump and filter. In the old process the platinum was deposited only on the surface of the porous nickel while in the new process the platinum is deposited throughout the porous nickel.

3.2 Effect of Bed Thickness

One method of adding additional water to the fuel cell and thereby increasing its capacity is to increase the thickness of the electrolyte bed. Before the modification could be applied to the final units, however, there are two questions that must be answered. The first question is that of the effect of the increased bed thickness on the voltage-current characteristics and the second is the effect of increased bed thickness on capacity.

In order to resolve these questions two experiments were initiated with small laboratory cell models. In the first experiment the voltage-current characteristics of the cell were measured with bed thicknesses of 1/32 inches (the currently employed bed thickness) and also 3/32 inches. The results showed that the discharge voltage-current characteristics for the cell

with larger bed thickness were slightly lower than those for the cell with smaller bed thickness, i.e., approximately 10-15 percent lower voltage at a given current density. This result signifies that although the use of a thicker electrolyte bed may give additional capacity, it would do so at a small reduction in power efficiency.

In order to resolve the second question the capacities of cells containing beds of various thicknesses are being measured. The end objective of these tests is to establish a plot of capacity versus bed thickness. The tests have not been completed at the writing of this report.

3.3 Palladium Catalyst

The feasibility of employing palladium as a substitute for platinum as the electrode catalyst was evaluated in a short-term investigation. The results indicated that palladium is just as effective as platinum for the O_2 electrode and that the amount required is less than for platinum. The results also indicated that mixtures of platinum and palladium on the H_2 electrode are just as effective as pure platinum. Since palladium is appreciably less expensive, a significant reduction in catalyst cost would be attained by using the electrodes employed in the second test.

3.4 Spacer Material

The high temperature encountered in the sterilization test limits the choice of plastics that may be employed as the cell spacer material. Testing of the currently employed Plexiglass "55" in a simulated sterilization test indicated that this material could not withstand the conditions, i.e., the high temperature causes excessive deformation of the spacer and "lifting" of the metal plating.

As a result of this test, it was decided to begin a search for a new material which could withstand the conditions of the test. Samples of several other types of plastics were subsequently received and evaluated. These materials included a polyester type resin, Sierracin 880", an acetal; Delrin, a polycarbonate: "Lexon", a phenolic "Nema Grade C", and a melamine; "Nema Grade G-5". Of these

several types, the most suitable was found to be the melamine plastic, i.e., the deformation was negligible and the metal did not lift from the plastic.

Before the melamine could be specified for the final units however, additional tests would have to be performed. These tests should include the machining of a sample spacer from this material (the above tests were conducted on small rectangular samples) and employing it in the cell assembly. The purpose of these tests would be to insure that the use of this new material will introduce no new variables.

4. DESIGN CALCULATIONS FOR FINAL UNITS

The heat generation and heat transfer relationships which were derived in the first quarterly report were applied to design calculations for the final units. The results of these calculations are given below. Also included is a section on the thermal capacity of the fuel cell.

4.1 Voltage-Heat Characteristics

The relationship between heat generation density and operating single cell voltage (for both charge and discharge) was given in the first quarterly report. The corresponding heat generation-voltage relation for a bank of 38 series connected cells, each with an electrode area of 28.3 in^2, was calculated on the basis of the above single cell data and is shown graphically in Fig. 2. Inspection of the charge curve indicates that heat generation is directly related to the polarization characteristics of the cell. The discharge curve indicates that internal losses increase rapidly with load, thereby significantly lowering cell efficiency.

4.2 Voltage-Power Characteristics

In addition to defining the heat generation rate, the operating voltage also defines the cell's power output (or input) by virtue of the cell's characteristic voltage-current curve. The voltage-current characteristic for a single cell is shown in Fig. 3, and the corresponding characteristics for the bank of 38 cells are shown in Fig. 4. The voltage-power characteristics for the bank of 38 cells is readily determined by calculating the voltage-current product from Fig. 4, the results of which are shown in Fig. 5.

Inspection of the charge portion of the curve in Fig. 5 indicates a near linear relationship between input power and terminal battery voltages greater than 60 volts. Inspection of the discharge

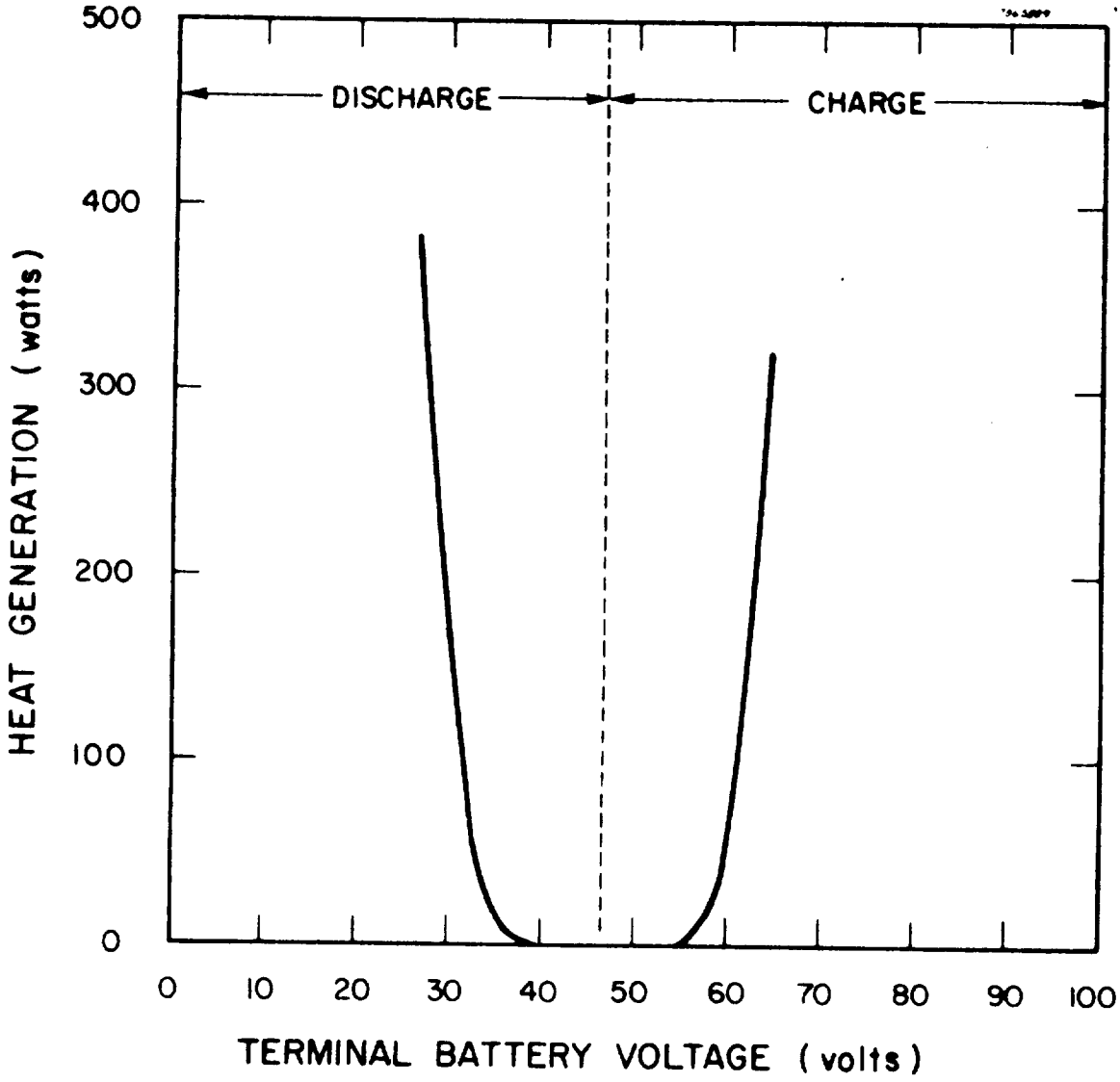


FIG. 2 VOLTAGE-HEAT CHARACTERISTIC FOR 38 CELL UNIT

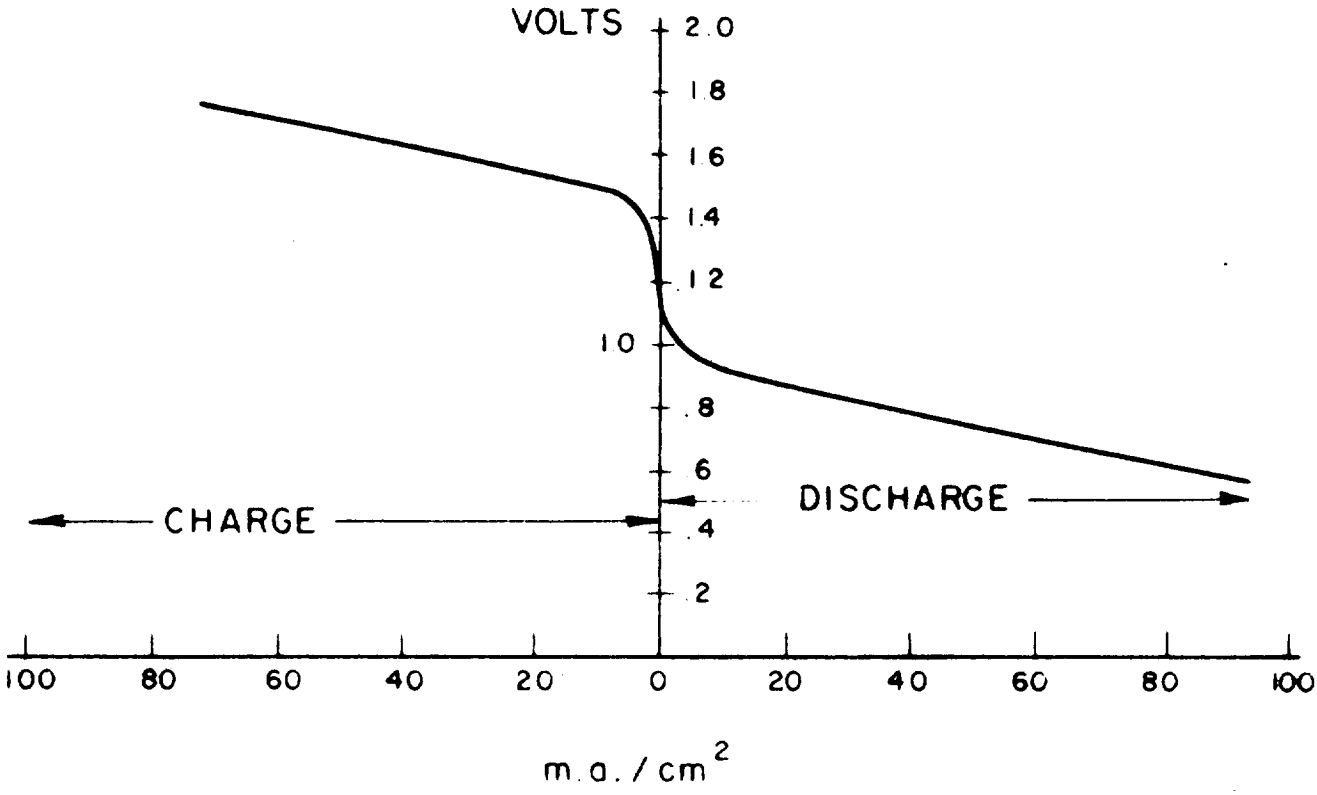


FIG. 3 VOLTAGE-CURRENT CHARACTERISTICS FOR SINGLE CELL

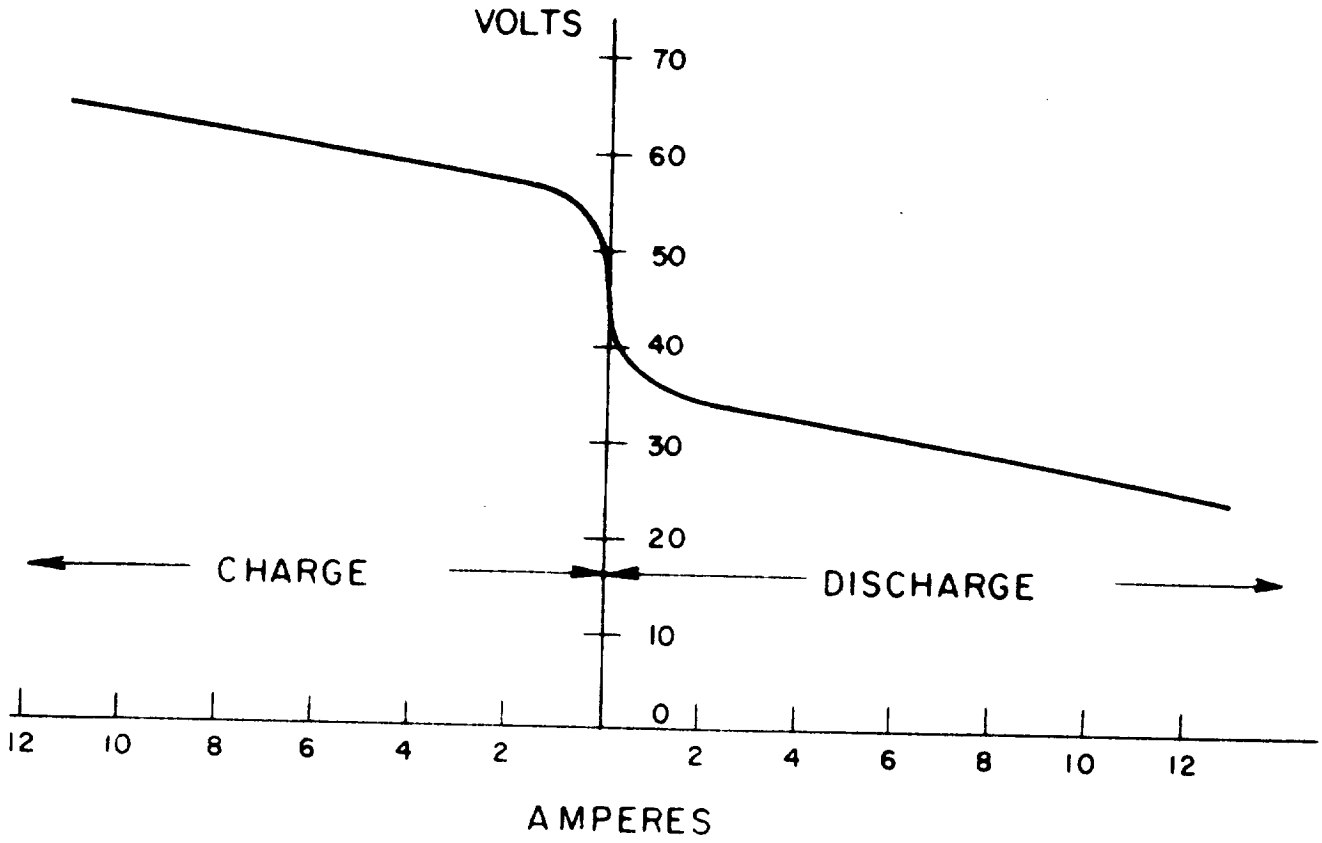


FIG. 4 VOLTAGE-CURRENT CHARACTERISTICS FOR 38 CELL UNIT

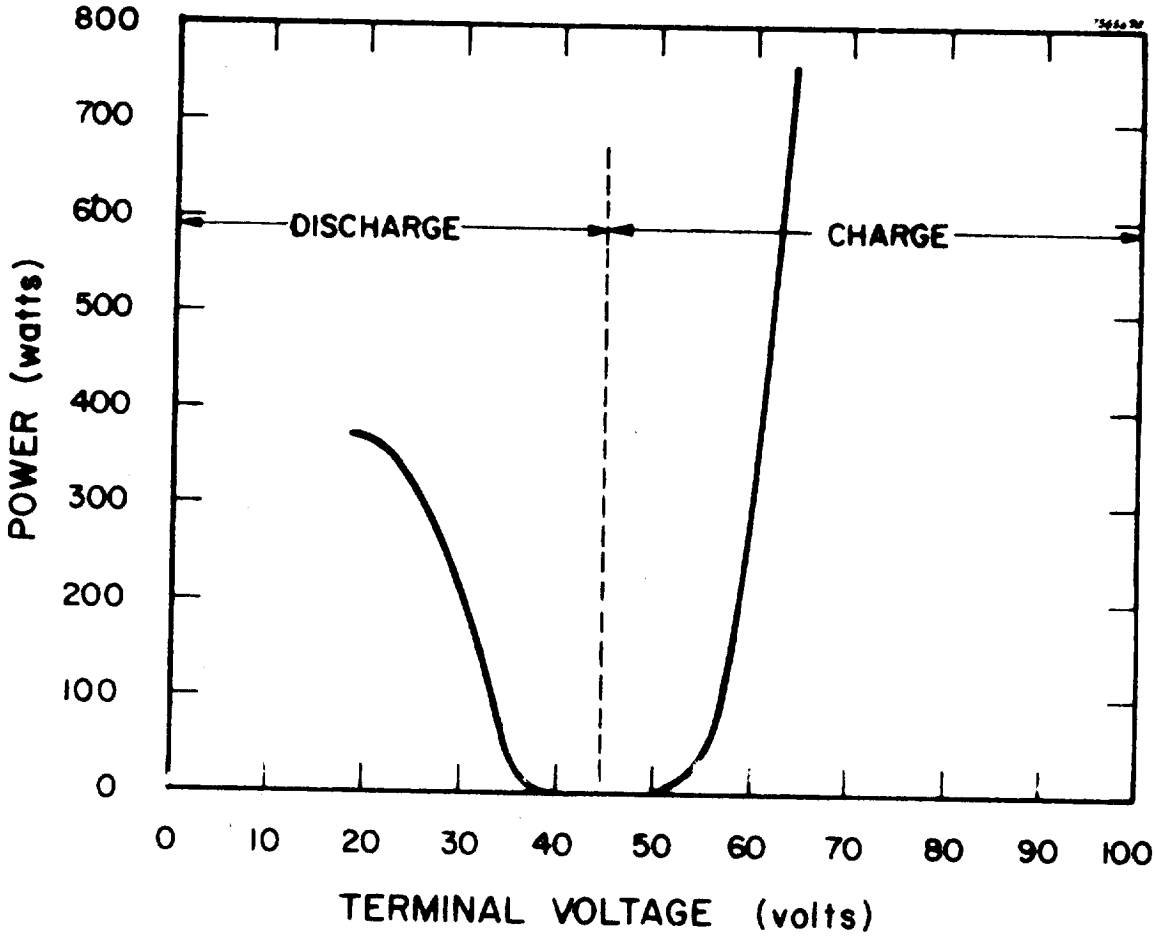


FIG. 5 VOLTAGE-POWER CHARACTERISTICS FOR 38 CELL UNIT

portion of this curve indicates that a maximum output power exists with respect to voltage at approximately 19 volts.

4.3 Heat-Power Characteristics

Since the operating cell voltage defines both the heat output and electrical output (or input), then there exists a relationship between heat and electrical output. With the voltage-heat data from Fig. 2 and the voltage power data from Fig. 5, the heat-power characteristics were tabulated and are shown in Fig. 6 for charge and Fig. 7 for discharge. Inspection of the curves presented in these figures indicates a parabolic relationship between heat generation and electrical power.

4.4 Temperature-Load Characteristics

In order to determine the relationship between internal cell temperature and electrical load (for continuous operation), it is necessary to know the thermal environment as well as the materials of construction and geometry of the fuel cell gas-cylinder assembly.

For the following design calculations, the thermal environment was assumed to be such that one-half of the assembly was exposed to black space and the other half was exposed to a surface (on the spacecraft) at 80°F.

The proposed geometry of the fuel cell gas cylinder assembly as well as the materials of construction were all outlined in the first quarterly report . A schematic diagram of the proposed assembly and three modifications thereof are shown in Fig. 8. The four assemblies are identical except in the number of locations of base plates. Figures 8-A and 8-B consider the use of an end and a center-mounted base plate respectively. Figure 8-C considers the use of two base plates each of which is located at a distance of 1/4 "L" (where "L" is the length of the stack of fuel cells) from the end of the assembly. Figure 8-D considers the use of three base plates, one of which is located in the middle of the cell stack, and two which are located at a distance of 1/6 "L" from either end of the cell stack.

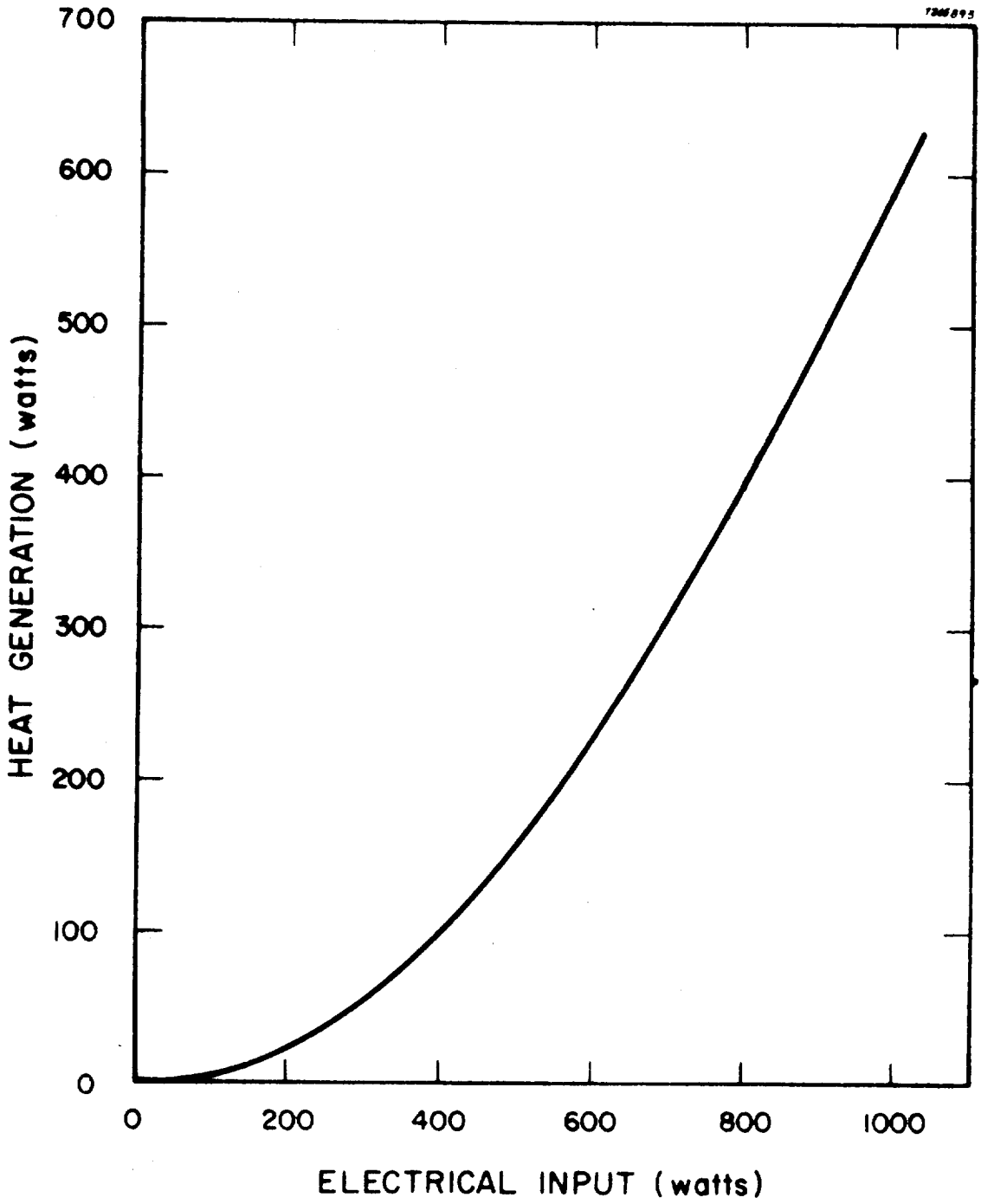


FIG. 6 HEAT-POWER CHARACTERISTICS OF 38 CELL UNIT ON CHARGE

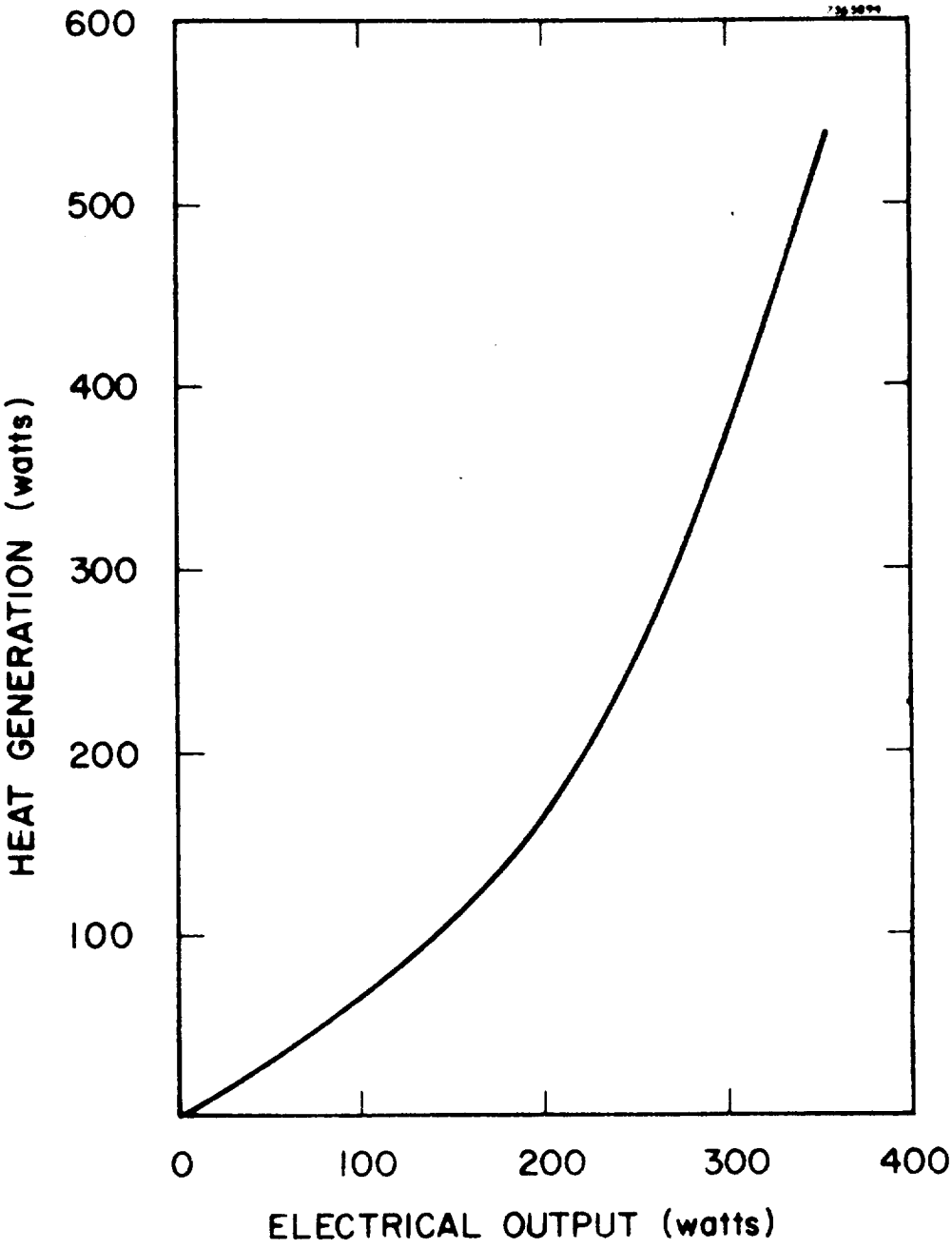
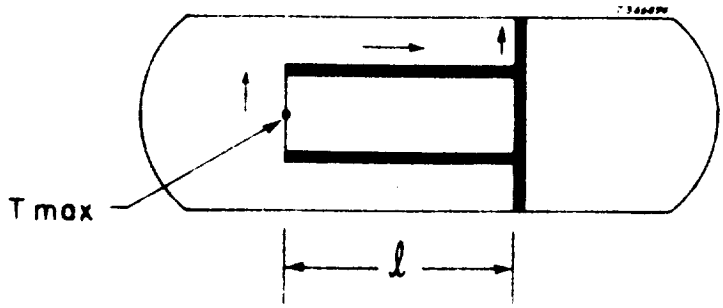
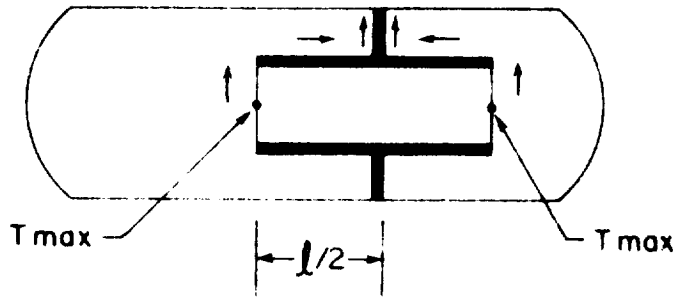


FIG. 7 HEAT-POWER CHARACTERISTICS OF 38 CELL UNIT ON DISCHARGE



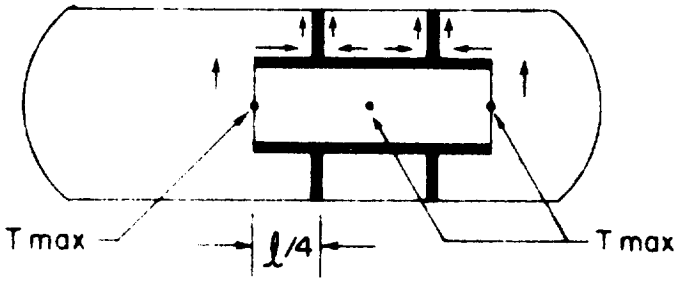
CASE 1

ONE END MOUNTED
BASE PLATE



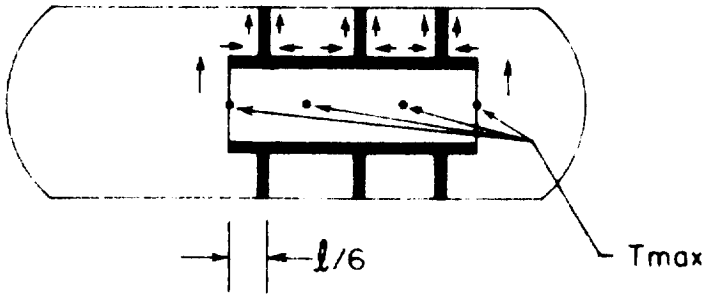
CASE 2

ONE CENTER MOUNTED
BASE PLATE



CASE 3

TWO BASE PLATES
EACH 1/4"l" FROM END



CASE 4

THREE BASE PLATES
ONE IN CENTER AND
TWO EACH 1/6"l"
FROM END

FIG. 8 PROPOSED DESIGNS FOR FINAL UNITS

The sheath and base plate were assumed to be made of 1/2 inch thick aluminum and the gas cylinders were assumed to be 1/4 inch thick aluminum. The length of the sheath is 12 inches and the diameter of the base plate is 15 inches. The fuel cell spacers are 8 inches in diameter and contain a metallic coating which consists of 3 mils of copper and 3 mils of nickel.

For the above geometry, materials and thermal environment, the internal cell temperatures may be determined as a function of the electrical load according to the procedures outlined in the last quarterly report.

The cell temperatures within the stack will not be equal however, and there will exist a gradient from one cell to the next. The location of the maximum temperature within the assembly will be approximately at the center of that cell which is farthest away from the base plate. For the first case, there will be one point of maximum temperature on the cell at the extreme left of the assembly (see Fig. 8-2). For the second case, there will be two points of maximum (and equal) temperature located on the cells at the extreme left and right of the assembly. For the third case, there will be three points of maximum (and equal) temperature located on the cells at the extreme left and right as well as the one in the middle. Finally, in the fourth case, there will be four points of maximum (and equal) temperature two of which are located on the extreme left and right and two of which are located at a distance 1/3 "L" from either end.

For continuous operation at high current efficiencies it is essential that the internal cell temperature not exceed 200°F. Therefore, if this temperature is taken as the upper operating limit, the curves in Figure 10 signify that the maximum electrical output is between 225 and 275 watts (depending on the number and location of base plates). Further examination of these curves indicates that a significant reduction in temperature can be accomplished by merely changing the location of a single base plate from the end to the center of the cell stack.

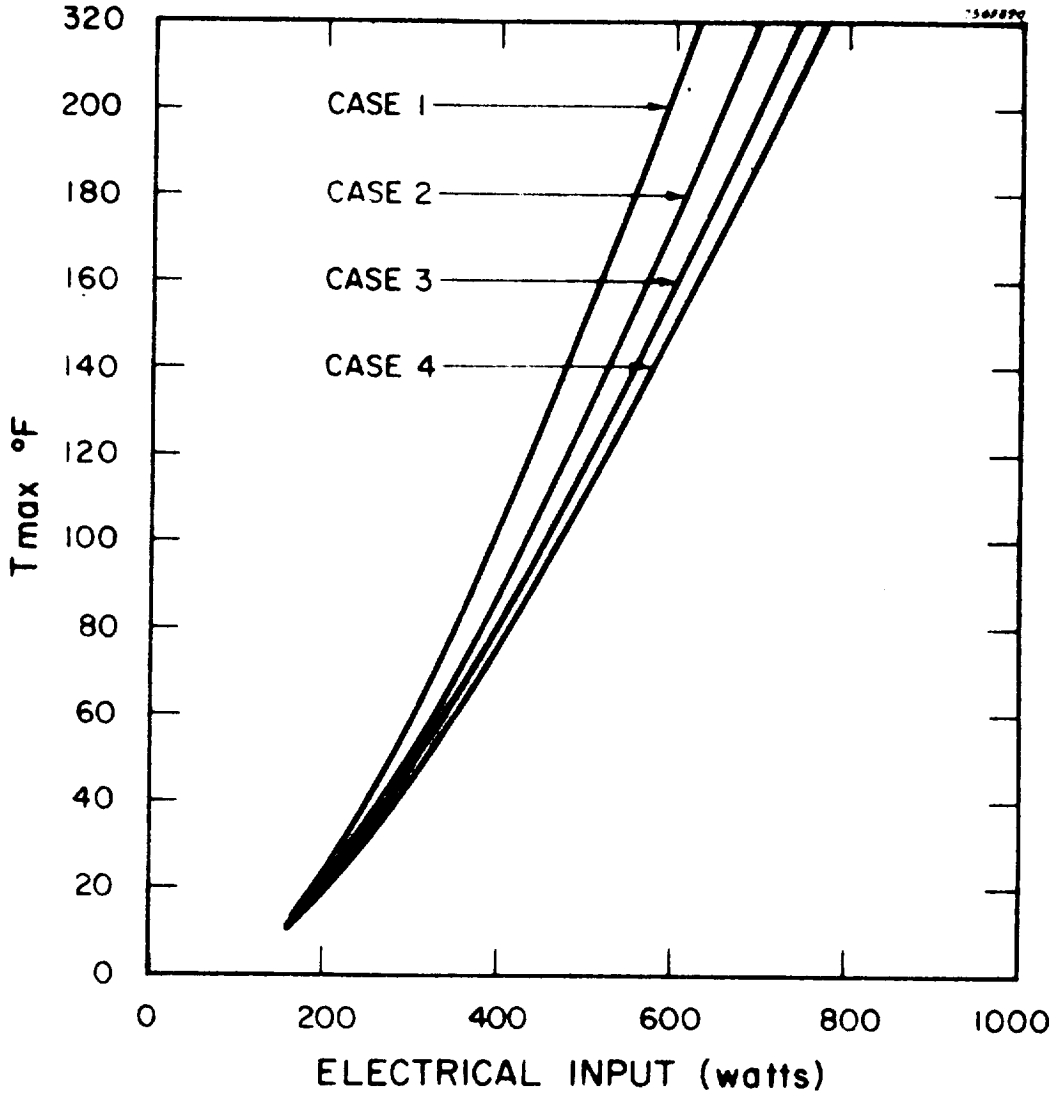


FIG. 9 MAXIMUM TEMPERATURE ON CHARGE

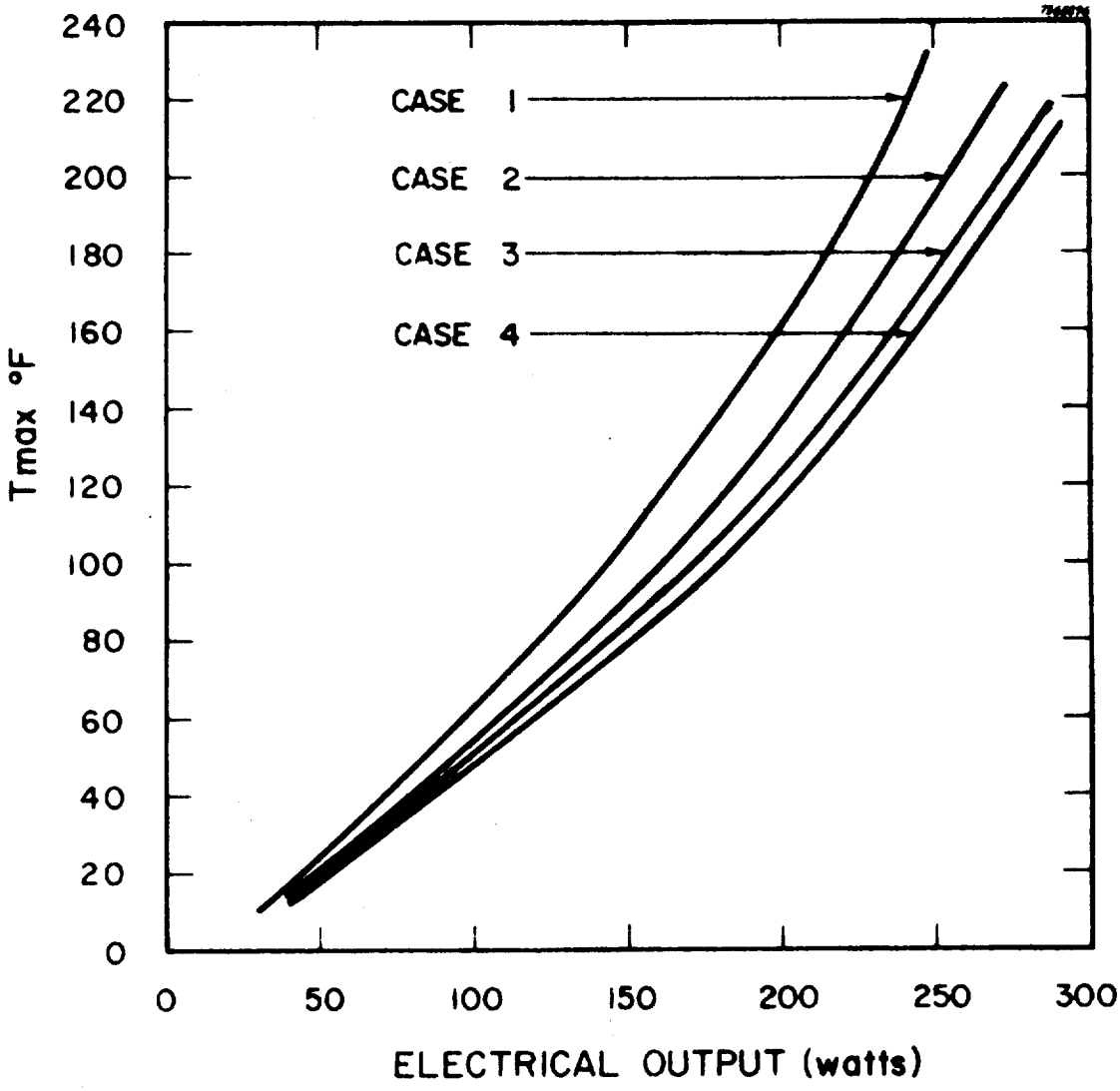


FIG. 10 MAXIMUM TEMPERATURE ON DISCHARGE

96

4.5 Thermal Capacity Characteristics

If conditions were such that the fuel cell in either of the above units were to operate under adiabatic conditions, then the operating power and time would be limited by the thermal capacity of the fuel cell.

The thermal capacity of the fuel cell components as well as the total capacity for the bank of cells is tabulated below. The weights of the materials are for a bank of 38 cells. The metals include the nickel electrodes as well as the plating on the cell spacers. The plastic is polymethyl-methacrylate and is used for the spacers. The rubber is buna-N and is used for the "O" rings.

Material	Weight gms	Sp. Ht. cal/gm/^{o}C	(Wt) x (Sp. Ht.) cal/^{o}C
		Thermal Capacity	
Plastic	6380	0.35	2240
Rubber	342	0.45	154
Metal	3790	0.10	379
Asbestos	1140	0.25	285
KOH Soln	760	0.70	531
TOTAL	12,612		3589

The total thermal capacity of the fuel cell is thus found to be 3589 cal/^{o}C. In other units, this may be expressed as 7.9 BTU/^{o}F or 2.32 wh/^{o}F.

Based on this thermal capacity, the rate of temperature rise was calculated for various electrical outputs. With the aid of this analysis, the maximum operating time may be calculated for various power levels. For example, if the cell were initially at room temperature, 70^{o}F, and were discharged at 100 watts, the time required for the

internal temperature to reach 200°F is 5.2 hrs. Since the maximum operating temperature is 200°F, then the maximum operating time is 5.2 hrs. Similarly, at an output of 200 watts, the operating time is found to be approximately 2 hrs.

5. SAFETY CONSIDERATIONS

In view of the fact that this system contains the potentially explosive gases, hydrogen and oxygen, some consideration was given to the possible safety hazards that may be involved in the operation and handling of the prototypes and units. The results of this analysis indicate that although the possibility of a H_2-O_2 explosion cannot be ruled out altogether, the chances for such an occurrence can be greatly diminished by proper design. The results also indicate that should such an explosion occur, the internal pressure rise would be 6.6 times the initial gas pressure. Therefore, in order to design a unit that would be completely safe even in the event of an explosion it would be necessary to specify a maximum operation pressure of 6.6 times the maximum pressure in the fully charged state.

6. PROBLEM AREAS

One of the major problems that has yet to be resolved is that of devising a method for testing the electrochemical activity of the platinized electrodes. The need for such a method is readily apparent, since the insertion of an inactive electrode within the cell stack causes high cell polarization and may lead to cell reversal on load. The present method for preventing the above phenomena is to discard the electrode altogether and insert a new one. Although some methods are known for testing the activity of catalysts for other uses, the results are not necessarily applicable to the measurement of electrochemical activity.

Another problem which has been encountered is that of uniformity of the asbestos stack material. Thickness variations of $\pm$ 10 to 15% have been found within a given lot and there are some indications that the chemical composition may vary from lot to lot. Since both of the above factors can effect cell performance, it will be necessary to engage in a more critical quality control inspection of this raw material.

7. CONCLUSIONS

Assembly and operation of a stack of at least 34 series connected cells has been experimentally demonstrated.

The capacity of the first prototype is at least 20 amp-hrs.

Thorough flushing of the hydrogen and oxygen cylinders with their respective gases is an essential part of the assembly procedure.

Improved electrode performance can be obtained by a modification of the platinization procedure.

Palladium is an effective substitute for part of the platinum on the hydrogen electrode and completely on the oxygen electrode.

The thermal contact resistance between the electrodes and the cell spacers is tolerably low for the designed heat loads.

A melamine plastic is the most suitable high temperature material for the cell spacers, if sterilization temperatures of 150°C become a future requirement.

The maximum pressure that can develop in a hydrogen-oxygen explosion is approximately 6.6 times the initial gas pressure. The cylinders of the first prototype have a small margin of safety when charged to the 25 amp-hr level.

A suitable design for the final units would be similar to the design of the first prototype, but may contain a center mounted base plate. The thermal capacity of the fuel cell must be taken into account in heat transfer design for short cycles.

8. PROGRAM FOR NEXT QUARTER

The primary emphasis during the initial portion of the next quarter will be devoted to the testing of the first full-scale unit. These tests will include the measurement of the voltage-current characteristics as a function of temperature and pressure and the measurement of voltage, and temperature during constant current cycling. Charge retention tests will also be conducted.

Redesign and fabrication of the two additional units will proceed immediately upon the completion of the above tests. Cycling of the first unit will be continued during the fabrication of the second and third units.

Additional problems have been encountered with the gas storage tanks which are required for testing of the complete multi-cell assembly in the true secondary mode. The sealing flanges, which contain the O-ring glands, were deformed during hydrostatic proof testing of the tanks. A redesign, incorporating high strength flanges, has been accomplished and a new assembly is in fabrication with delivery anticipated by 31 October 1962.